Structural, Dielectric and Electrical Properties of $Sr_{1-X}Zn_XTiO_3$(x=0.5) Ceramics

K.Chandra Babu Naidu, T.Sofisarmash and T.Subbarao

Abstract: The present study deals with structural, morphological, dielectric and electrical properties of Zinc doped $SrTiO_3$ powder particles which have been prepared via conventional solid-state route method. These were ball milled for 8 hr and furthermore calcined at 1050^0C for 8hrs and sintered at 1250^0C for 2hrs.The dielectric constant (K), dielectric loss (tanδ) and thermoelectric powers (S) were computed. The obtained results using HIOKI 3532-50 LCR HiTESTER showed that the sample exhibited high dielectric constant of 246.6 at RT (100 Hz) and low loss of 0.50195 at RT (10 kHz).The samples were characterized using XRD, SEM,FTIR, EDAX and TEP for structural, micro structural, functional group, elemental and thermoelectric power analysis respectively.

Keywords: Solid-state route method; thermoelectric power; dielectric constant; XRD; SEM;EDAX;FTIR.

1 INTRODUCTION

Zn doping induces the dielectric response of quantum Para –electric $SrTiO_3$ [1].Also exhibits the photo catalytic activity with double time improvement than the undoped $SrTiO_3$ [2].Hence Zn doped $SrTiO_3$ samples have got extensive applications in microwave devices, resonators, tunable capacitors, phase shifters and oscillators owing to their high dielectric constant and low loss at room temperature. The substitutional doping of appropriate ions could change the crystal and band structures and significantly can promote electron transport in catalysts due to their good photo catalytic activities. Zinc doped strontium titanate can also reduce the surface defects of the undoped strontium titanate (ST). $SrCO_3$, $ZnCO_3$ and TiO_2 were taken as the raw materials of 99.9% purity in their appropriate proportions and consequently the final powders were characterized. Dielectric constant (K), dielectric loss (tanδ), ac-conductivity (σ_{ac}) and thermoelectric power (S) were investigated. Dielectric loss quantifies a dielectric material's inherent dissipation of electromagnetic energy into, e.g., heat [3]. It can be parameterized in terms of either the loss angle δ or the corresponding loss tangent tanδ. In the recent investigations the photo catalytic activity, structural, surface morphological, Raman spectrum, dielectric and UV-Visible studies have been revealed [1,2].But in this study the author intended to deal with the structural, micro structural dielectric, thermoelectric and conductivity properties of Zn doped $SrTiO_3$. Thermoelectric properties possessed materials have got extensive applications in power generation (solar thermal generation) [4] and refrigeration. This is achieved by means of seebeck effect which converts the heat energy into electrical energy. The well fitted thermoelectric material is bismuth telluride. However zinc doped strontium titanate also exhibits these properties in a good level i.e. in terms of micro volts per Kelvin electrical energy is being produced. Thermoelectric efficiency depends on the figure of merit, ZT.

There is no theoretical upper limit to ZT, and as ZT approaches infinity, the thermoelectric efficiency approaches the Carnot limit. However no known thermoelectric materials have a ZT >3 [5].

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2 PREPARATION OF THE SAMPLE

$Sr_{1-X}Zn_XTiO_3$(x=0.5) powder particles have been prepared via conventional solid state reaction method. In order to prepare the Zn doped $SrTiO_3$ particles initially $SrCO_3$, $ZnCO_3$ and TiO_2 were taken as the raw materials of 99.9% purity. Subsequently SrCO3, $ZnCO_3$ and TiO_2 were incorporated into fresh crucible and kept in a furnace which can enable to go up to the temperature 1300^0C .Hence $Sr_{1-X}Zn_XTiO_3$(x=0.5) powder particles were prepared. The obtained Zn doped $SrTiO_3$ particles were grinded by making use of agate mortar into a fine powder. Later they were ball milled for nearly 10hrs and calcined at a temperature 1050^0C in air for nearly 8hrs.Afterwards the calcined sample is cooled to room temperature at the rate of 5^0C/min. The formed compound is again grinded and pressed into pellets by adding a binder PVA using pellet machine with an application of pressure ranging from 10 to 20 tons. Furthermore the pellets prepared were undergone sintering at a temperature 1250^0C for 2hrs.The pellets were coated with silver paste on both sides. Afterwards the samples were characterized by X-ray diffract meter (XRD), Scanning electron microscope (SEM),and Energy dispersive X-ray spectrometer(EDAX) for structural and micro structural, elemental composition analysis respectively. Not only these but also characterized by HIOKI 3532-50 LCR HiTESTER and TEP measurement kit for

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measuring dielectric constant, dielectric loss ,quality factor, ac conductivity and seebeck-coefficient(thermo electric power).Apart from these dc-conductivity and FTIR analysis have also been performed for finding dc-activation energy and functional groups present in specimen

3 RESULTS AND DISCUSSIONS.

3.1 Dielectric Properties

Dielectric constant (K), dielectric loss ($\tan\delta=Q^{-1}$), dielectric relaxation, dielectric tunability and polarization were regarded as the dielectric properties. In $Sr_{1-x}Zn_xTiO_3$(x=0.5) only dielectric constant and dielectric loss were investigated. Apart from these properties quality factor (Q) and ac conductivity (σ_{ac}) were also computed using HIOKI 3532-50 LCR HiTESTER .The obtained results showed that the dielectric constant is increasing with increase of temperature and decreasing with increase of frequency. The sample exhibited high dielectric constant of 246.6 at RT (100Hz) and 1507.42 at 400^{0}C (100Hz).The loss is increasing with increase of temperature and decreasing with increase of frequency. The sample exhibited low loss of 0.50195 at RT (10 kHz). The high dielectric constant at room temperature provides applications in microwave communication systems, resonators and capacitors needed circuitry systems. The substitution of Zn into the quantum paraelectric $SrTiO_3$ improved the dielectric nature of the present compound [1]. The dielectric constant (K) and ac conductivity (σ_{ac}) have been measured using the following formula.

$$K=C/C_0 \text{ or } Cd/\varepsilon_0 A$$

The following plots show the variation of dielectric constant (K) Vs temperature and dielectric loss($\tan\delta$) Vs temperature, of $Sr_{1-x}Zn_xTiO_3$(x=0.5).

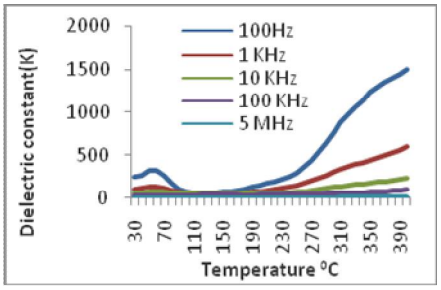

Fig1.shows the variation of dielectric constant Vs temperature of $Sr_{1-x}Zn_xTiO_3$(x=0.5)

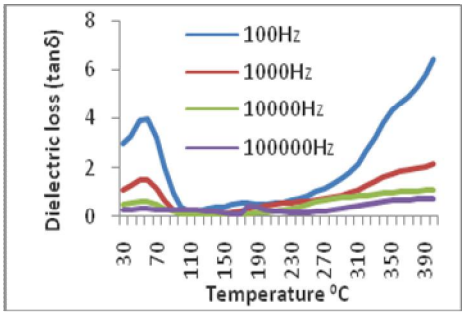

Fig2.shows the variation of dielectric loss Vs temperature of $Sr_{1-x}Zn_xTiO_3$(x=0.5)

3.2 AC-Conductivity

The ac conductivity is increasing with increase of temperature. The following plot shows the variation of ac-conductivity (σ_{ac}) Vs temperature of $Sr_{1-x}Zn_xTiO_3$(x=0.5). AC- conductivity is increasing with the increase of temperature. However at high temperatures there is an abrupt increase in AC conductivity which may be due to strong hopping mechanism.AC-activation energy (E_a) also computed as $1.088X10^{-3}$eV. AC conductivity (σ_{ac}) and AC-activation energy (E_a) were calculated using the following formulae.

$$\sigma_{ac}= K\ \varepsilon_0\ \omega\ \tan\delta$$

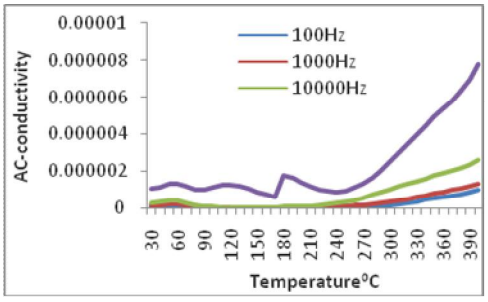

Fig3.shows the variation of ac conductivity Vs temperature of $Sr_{1-x}Zn_xTiO_3$(x=0.5)

3.3. DC-conductivity

DC-conductivity ($\sigma_{dc}=L/RA$) of $Sr_{1-x}Zn_xTiO_3$(x=0.5) was performed and it exhibited the increasing nature with respect to the temperature from RT to 400^{0}c. Apart from this $\ln\sigma$ Vs 1000/T plot has been also performed in fig.5.From the plots drawn natural logarithm of dc-conductivity ($\ln\sigma$) against the reciprocal of temperature (1000/T) activation energy (E_a) of Zn doped $SrTiO_3$ ceramic material was found as 0.159 eV using the following formula. In the graphs shown the activation energy and conductivity are increasing with increase of temperature and it is observed that the activation energy is very low at RT. It may be due to generation of charge carriers and mobility due to raising temperatures.

$E_a=2K_b\ln\sigma/(1000/T)$ Where $K_b=8.86X10^{-5}$eV (Boltzmann constant)

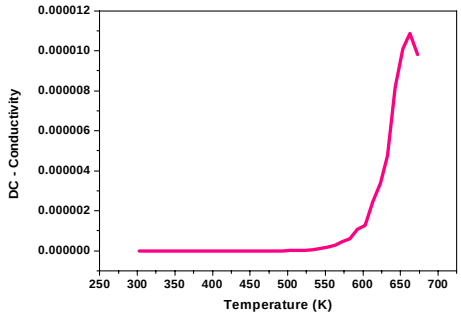

Fig4.Shows σ_{dc} Vs Temperature Plot of $Sr_{1-x}Zn_xTiO_3$(x=0.5)

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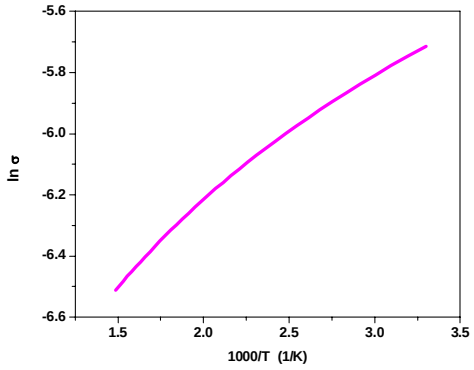

Fig 5.Shows lnσ Vs 1/T plot of $Sr_{1-x}Zn_xTiO_3$(x=0.5)

3.4 Thermoelectric Properties

The thermoelectric properties of $Sr_{1-x}Zn_xTiO_3$(x=0.5) have been studied by measuring the seebeck coefficient. At temperature 308^{0}K the present sample exhibited thermoelectric power of 6800μV/^{0}C. The thermoelectric power behavior of Zn doped perovskite $SrTiO_3$ is shown in the plot. As increasing the value of temperature, the seebeck coefficient is decreasing. At room temperature the specimen performed high seebeck coefficient. Due to the incorporation of Zn into the lattice of strontium titanate exhibited better improvement than undoped $SrTiO_3$ as far as the thermoelectric power is concerned. These materials are the good targets for the materials researchers. Not only this, but for wide-spread commercial applications such as the motor industry, the materials used must be environmentally friendly and non-toxic.

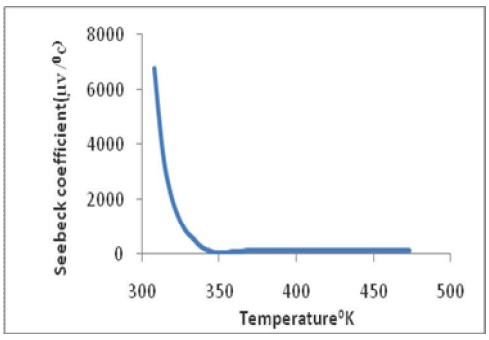

Fig.6 shows the variation of temperature Vs seebeck coefficient of $Sr_{1-x}Zn_xTiO_3$(x=0.5)

3.5 FTIR analysis

FTIR is mainly for the elucidation of functional groups present in a sample. In the present investigation of the sample $Sr_{1-x}Zn_xTiO_3$(x=0.5) the transmittance spectrum is performed. It exhibited the graph as shown below. This information provides both qualitative and quantitative analysis of organic and inorganic compounds.The transmittance spectrum of present sample called the $Sr_{1-x}Zn_xTiO_3$(x=0.5) acquired broad bands at the wave numbers 625.62247 cm^{-1} and 817.757920 cm^{-1} due to the presence of Ti-O stretching vibrations [6] and the other band at wave number 1508.377512 cm^{-1} may probably due to presence of Zn-O stretching vibrations. As the sample is of ceramic type no organic functional group was present.

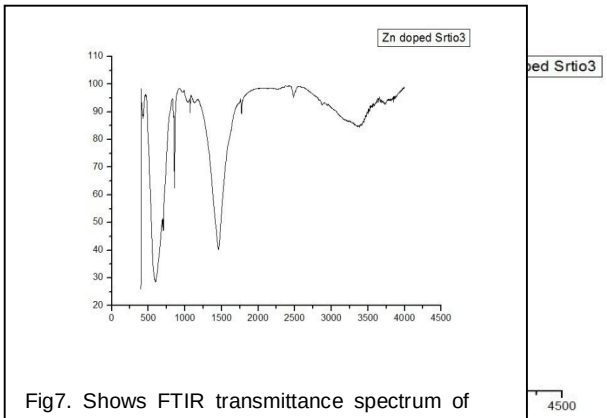

Fig7. Shows FTIR transmittance spectrum of $Sr_{1-x}Zn_xTiO_3$(x=0.5)

3.6 XRD analysis

$Sr_{1-x}Zn_xTiO_3$(x=0.5) powders were analyzed for crystalline structure by X-ray diffractometry (XRD, Rigaku) using Cu K_α (0.15418nm) radiation in order to identify the phases formed after being heat treated. The average crystalline size (D_p) was calculated according to the Scherrer's formula. Moreover the average dislocation density (ϱ) and average elastic strain (E_{strain}) were computed. The formulae related to D_p, ϱ and E_{strain} are as follows.

$$D_p = K\lambda/\beta \cos\theta$$

Where K is a constant and is equal to 0.9 &

λ=0.15418 nm.

$$\varrho = 1/D^2 \text{ and}$$

$$E_{strain} = \beta/4\tan\theta$$

Where β is full width half maxima.

Fig 8. Shows the XRD pattern of $Sr_{1-x}Zn_xTiO_3$(x=0.5) powders calcined at 1050^{0}C. There are some Intermediate phases formed in XRD spectrum in addition to the peaks concerning to the XRD spectrum of Pure $SrTiO_3$.These extra peaks may be due to the presence of $ZnTiO_3$ [7]. Since the concentration of Zinc equals to the concentration of Strontium, intensity of intermediate phases is high. The miller indices were (100),(110),(111),(200),(210),(220),(300),(310),(222),(023),(004) represented in fig.8 and lattice parameters were computed as 4.87018 (a=b=c) and 90^0 ($\alpha = \beta = \gamma$). Hence the structure of sample $Sr_{1-x}Zn_xTiO_3$(x=0.5) is cubic. The average crystalline size, average dislocation density and elastic strain were noted as 109 nm, 0.842x10^{-15} m^{-2} and 0.26931 respectively. In XRD spectrum the intensity of

the $ZnTiO_3$ peaks is high due to having 0.5 (high) concentrations in perovskite ST.

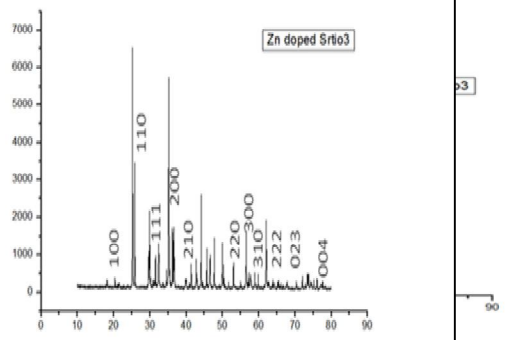

Fig8. Shows XRD spectrum of $Sr_{1-X}Zn_XTiO_3$ (x=0.5)

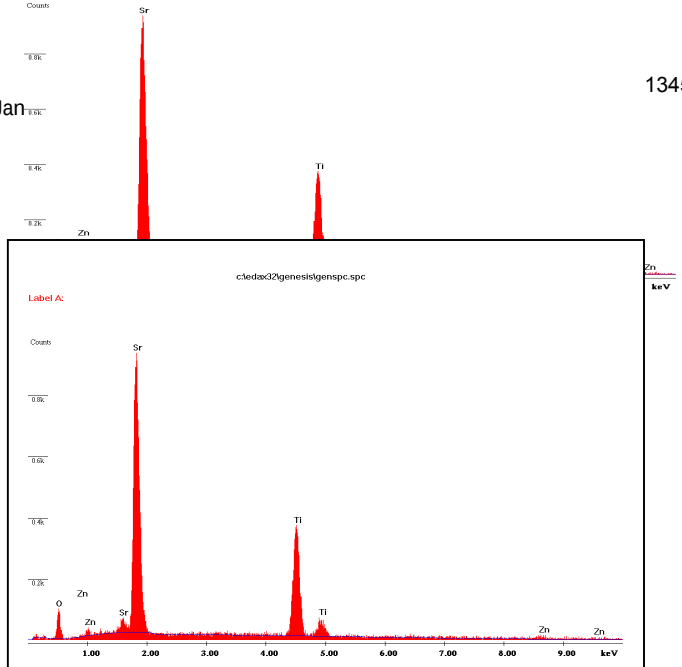

Fig10.Shows EDAX image of $Sr_{1-X}Zn_XTiO_3$(x=0.5)

3.7 SEM and EDAX analysis

This approach is useful in qualitatively or semi-quantitatively determining chemical compositions, crystalline structure and crystal orientations. The average grain size (G_a) of the sample $Sr_{1-X}Zn_XTiO_3$(x=0.5) calculated as 1.65µm using the following formulae. The surface morphology was studied at the spot having the magnification 16KX.

Average grain size G_a = 1.5 L/MN. Where L=the total test line length, M=the magnification, N=the total number of intercepts which the grain boundary makes with the line.

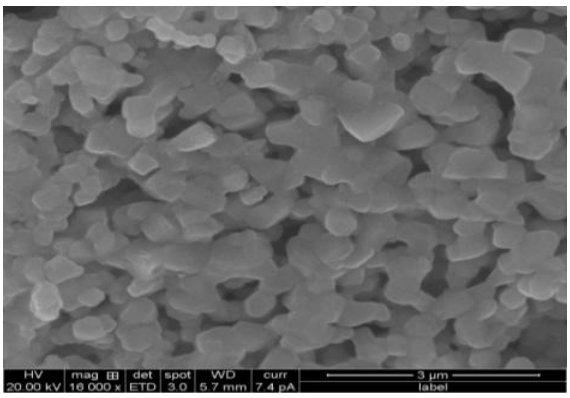

Fig 9.shows SEM image of $Sr_{1-X}Zn_XTiO_3$(x=0.5)

Energy Dispersive X-ray Spectroscopy can be used to find the chemical composition of material down to a spot size of a few microns and to create element composition maps over a much broader raster area.

Elem	Wt %	At %
O K	8.98	29.31
SrL	54.74	32.64
TiK	31.01	33.83
ZnK	5.27	4.22
Total	100.00	100.00

4 CONCLUSIONS

Strontium titanate with Zinc showed good thermoelectric, dielectric properties. The micro structure of the sample is observed at high sintering condition i.e. at 1250^{0}C.In the EDAX spectrum the maximum peak obtained for Sr. AC -conductivity (σ_{ac}) shows sharp increase due to linear hopping mechanism, dc conductivity (σ_{dc}) is decreasing with respect to the temperature. In the case of material say
$Sr_{1-X}Zn_XTiO_3$(x=0.5) it exhibited DC- conductivity greater value than AC –conductivity. However, in most of the ceramic materials this condition is proved. FTIR and EDAX described the presence of groups Ti-O, Zn-O and the elements Oxygen, Strontium, Titanium, and Zinc. In SEM figure larger number of grains and few grain boundaries are observed.

5 ACKNOWLEDGEMENTS

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